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Carbon-14 Bomb Pulse Dating

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Abstract:

Atmospheric testing of nuclear weapons during the 1950s and early 1960s doubled the concentration of carbon-14 in the atmosphere and created a pulse that labeled everything alive since 1955 as carbon moved up the food chain. The variation in carbon-14 concentration in time is well-documented and can be used to chronologically date all biological materials since the mid-1950s.

Keywords: Radiocarbon, bomb-pulse, carbon-14, dating, isotope forensics

Introduction

Traditionally, radiocarbon dating has been considered to be an archeological tool rather than a forensic one (See Radiocarbon Dating). Radiocarbon or carbon-14 (^{14}C) is produced naturally in the upper atmosphere by nuclear reactions between nitrogen and neutrons generated by cosmic ray interactions with atmosphere. Single carbon atoms in the atmosphere are chemically reactive and are quickly oxidized to carbon dioxide CO_2 . The atmospheric concentration of natural ^{14}C with respect to all carbon has remained relatively stable at about 1.2 parts per trillion over the past several thousand years. With a radioactive half-life of 5730 years, the radioactive decay of ^{14}C is minimal within the time periods of interest in medical forensic cases and applicable for traditional radiocarbon dating of samples over 300 years of age. Willard Libby was awarded the Nobel Prize in Chemistry in 1960 for the development of radiocarbon dating [1].

Atmospheric testing of nuclear weapons during the 1950s and early 1960s doubled the concentration of $^{14}\text{C}/\text{C}$ in the atmosphere (Figure 1) [2]. From the peak in 1963, the level of $^{14}\text{CO}_2$ has decreased with a mean life of about 16 years, not due to radioactive decay, but due mostly to mixing with large marine and terrestrial carbon reservoirs. The ^{14}C has not actually disappeared, it has simply moved out of the atmosphere. The temporal variations of artificially high levels of atmospheric radiocarbon have been captured in organic material world-wide and thus offer an opportunity to determine a date of synthesis for biomolecules. Since radiocarbon is

incorporated into all living things, this pulse is an isotopic chronometer of the past 60 years.

The atmospheric $^{14}\text{CO}_2$ curve depicted in Figure 1 is a northern hemisphere annual growing season average. It is constructed using several different data sets that used tree rings, recent plant growth, and direct atmospheric sampling to provide carbon samples [3-8]. Since there were relatively few geographic sources of bomb-pulse ^{14}C , the upswing and the peak values of the curve do vary with location around the globe [5,6]. However, since CO_2 is a gas, the pulse of $^{14}\text{CO}_2$ mixed in the atmosphere with all other CO_2 to produce a relatively homogeneous distribution of atmospheric $^{14}\text{CO}_2$ by the late 1960s [9].

The isotopic content of new plant growth reflects the atmospheric radiocarbon concentration. New leaves are produced in a matter of weeks while larger fruit and vegetables form over the period of a month or two. Herbivores lag the atmosphere slightly because their primary carbon source is on the order of months old. Omnivores and carnivores lag the atmosphere further because their carbon sources are another step removed.

Within organisms, tissues turn over at different rates so ^{14}C levels vary between tissues. The date of formation of a tissue or specific biomolecule can be estimated from the bomb-curve by considering these lags in incorporation and relating the ^{14}C concentration with the date [10]. Using an annual average of the carbon intake over a

growing season can account for much food chain lag and produce a usable curve (Figure 1). Caution must be exercised when dating an elevated sample since the pulse is double valued. Placing a sample on the ascending or descending side of the pulse can often be accomplished if other information is available.

Measurement of radiocarbon samples

Today most ^{14}C dating analyses are conducted using accelerator mass spectrometry (AMS), although there are still labs that use decay counting. AMS is much faster and generally more precise than decay counting since it measures differences in carbon atom mass and is not constrained to wait for atomic decay. AMS can also use smaller samples than decay counting, an important issue when analyzing evidence.

Sample preparation and measurement details vary among AMS facilities, depending on the type of sample to be analyzed and the design of spectrometer. Routine radiocarbon analyses using accelerator mass spectrometry (AMS) are performed on samples containing 0.5-1 milligram carbon. Samples as small as 20 μg carbon can be analyzed at some labs, but measurement uncertainties are larger. Nearly all AMS facilities that perform high precision dating follow these general procedures to minimize contamination from outside sources of carbon and reduce measurement backgrounds. Samples are dried completely and then combusted with excess oxygen to produce CO_2 . The CO_2 is purified

to remove water vapor, nitrogen, oxides of nitrogen, and oxides of sulphur. It is then reduced to graphite or elemental carbon on metal catalyst, often cobalt or iron powder.

Primary standards, secondary standards, and backgrounds are similarly processed to produce graphite, which is the form of carbon analyzed by the majority of AMS systems [13]. Graphite is the preferred form of carbon because it can be made easily at high purity and it produces intense negative ion currents. It is important to have consistent sample source material (e.g., all carbon graphite) because different molecules ionize with different efficiencies. A handful of gas accepting ion sources that take direct feed of CO₂ exist, but they are not typically used for high precision ¹⁴C analyses.

The precision of bomb-pulse dating depends on the ability to measure the ¹⁴C concentration in a sample and the slope of the curve. It is relatively easy to achieve 0.5-0.8% precision of the isotope measurement when analyzing recent full-sized samples. As the slope of the pulse flattens, the uncertainty in ¹⁴C analysis translates into a larger chronological uncertainty. When the slope was steep, the uncertainty was typically ± 1 year. Since 2000, that same measurement precision yields a chronological uncertainty of ± 2-4 years.

Soft Human tissues amenable to dating.

Most soft tissues exhibit relatively rapid carbon turnover. In a forensic context, the ^{14}C concentration in soft tissues can be used to determine an approximate date of an event, such as death of an unidentified body. Shortly after the start of the bomb-pulse [14], Broecker et al. (1959) [15] noted that bomb-curve radiocarbon concentrations in human tissues lag the atmosphere, relating to dietary issues and tissue turnover [16,17]. They documented a lag time of about 1.1 years for blood and 1.8 years for lung tissue [15]. Libby et al. (1964) further noted that ^{14}C concentrations in human brain tissue can lag atmospheric levels by only a few months [18]. Nydal et al. (1971) found good agreement in radiocarbon levels between samples of human blood and hair [19].

Most soft tissues decay quickly in the environment and are unsuitable for dating. Hair is a notable exception [20]. Hair tends to resist rapid decay and is often found with skeletal remains. Hair also records exposure to toxins and drugs and has been used to determine causes of death. Although individual strands of hair have little mass, the carbon content of hair is high and milligram-sized samples are relatively easy to attain. Although not widely used to determine date of death, bomb-pulse dating of hair has been used in several cases [21-23].

Bone and Cartilage

Bone is among the preferred samples for traditional radiocarbon dating. Bone's ability to resist rapid decay while containing a relatively high concentration of carbon makes it a

desirable material for traditional dating [24]. Traditional bone dating uses a collagen extraction to avoid potential complications with mineral exchange of carbonates in the environment. Collagen is a protein and is not affected by environmental carbonate exchange like the mineral component of bone.

Attempts to use bone and cartilage for bomb-pulse dating have not been very successful. Bone and cartilage do not lock carbon in an inert structure like hair. Bone and cartilage are alive and exhibit low but variable turnover, depending on physical activity, age, health, and type of bone [25-33]. Older individuals tend to lose more bone than they replace during the bone recycling process. In general, bomb pulse dating of bone can be used to determine if someone was alive during the period of the pulse, but cannot determine a date of birth or death. Cartilage has the same limitations as bone. In 1964, an autopsy study of the cartilage collagen of 70-year-old adults showed little increase of artificial radiocarbon despite living throughout the entire rise of the bomb-pulse from 1955 to 1964. [18].

Teeth

Although dental enamel is the hardest substance in the body, teeth are not routinely used in traditional radiocarbon dating due to fear of carbonate mineral exchange during centuries of burial. After being produced, there is no turnover of enamel, and the ^{14}C concentration reflects the level in the atmosphere at the time of enamel formation. Since teeth are formed at distinct, well-documented ages during childhood [33,34], the ^{14}C

concentration in dental enamel can be used to determine an approximate date of birth [35-42]. The absence of bomb-pulse carbon from enamel places date of birth in the 1940s or earlier. The technique reports accurate determination of teeth of known age with precision ± 1.5 years, a significant improvement over previous techniques.

Processing of enamel samples is different than soft tissues because the carbon resides in a mineral matrix. Enamel samples must be dissolved in acid and the liberated CO₂ must be trapped for isotopic analysis [38].

The live part of the tooth, principally dentin in the root, is like bone, with high collagen content and slow turnover and recycling of the carbon. It provides little direct information from ¹⁴C other than whether an individual was alive during the pulse [31]. Since the root is alive, its carbon is always younger than that of the enamel. Cook et al (2006) extracted collagen from dentin and showed it was younger than mineralized carbon in the enamel [36]. The mineral component of the root is also younger, possessing a different ¹⁴C/C ratio than the enamel. Knowing that the carbon in the root is younger than that in the enamel allows investigators to determine if a tooth was formed on the ascending or descending part of the bomb pulse with a single tooth. Enamel formation proceeds step-wise and careful dissection of enamel from a single tooth can yield statistically different ratios from different parts of the enamel during the time that the atmospheric ¹⁴C/C was changing more rapidly than it is today [41].

Efforts to develop a laboratory test for assessing age at death from teeth has focused on measurements of racemization of aspartic acid [43-48]. Aspartic acid slowly changes its structure over time at body temperature, but slows dramatically at cooler temperatures. Racemization analysis works best in cold climates where ambient temperature is well below body temperature. The technique does not work for bodies that have been burned at high temperature. By measuring the ratio of one orientation (D-aspartic acid) to the other (L-aspartic acid) an approximate age can be determined. Reported precision varies widely among labs over the past 30 years [43-48]. Recent studies report precision of ± 1.5 years, similar to bomb-pulse dating, but internal standards of identical age are required. A single tooth can be split and analyzed for both aspartic acid and ^{14}C to get estimates of date of birth and age at death [37]. Furthermore, stable isotope analyses of ^{13}C can provide geographic information about the residence of the individual during tooth formation [39].

Documents

Radiocarbon dating is routinely used to date archeological and art objects to the correct era. The purpose of the date is to detect forgeries. In theory, the same approach can be used to date documents originated during the bomb-pulse. Unfortunately, the carbon in paper is not from a specific year due to the relatively long lifetime of trees. The absence of bomb-pulse carbon in paper does not necessarily indicate a document is more than 50 years old. The presence of bomb-pulse carbon does however place the document after the onset of the pulse. Some glues contain large amounts of collagen which are usually

obtained from animal products. Since animal derived collagen is generally from animals only a couple years old, collagen from glue could narrow the age of glue to within a couple of years. Traditional photographs and photographic emulsions on negatives contain gelatin made from collagen. The collagen could also be used date photographs and negatives in the photographic art arena [49]

Seeds, Pollen, and Agriculture

Seeds and pollen are used in traditional radiocarbon dating to place archeological sites in a historical context. Since both are produced during a relatively short window of time in a single growing season, they are considered good archeological dating samples.

Because Seeds and pollen are produced during limited times, they are potentially useful for placing forensic evidence in a historical context. The bomb pulse has been used to date agricultural products in a couple applications [50]. Vintages of recent wines can be confirmed [51]. Interdicted contraband, such as illegal drugs, can be dated with the pulse to determine year of production. Products such as ivory from endangered species can also be dated [52], but customs services have not routinely used the technique.

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Fig. 1

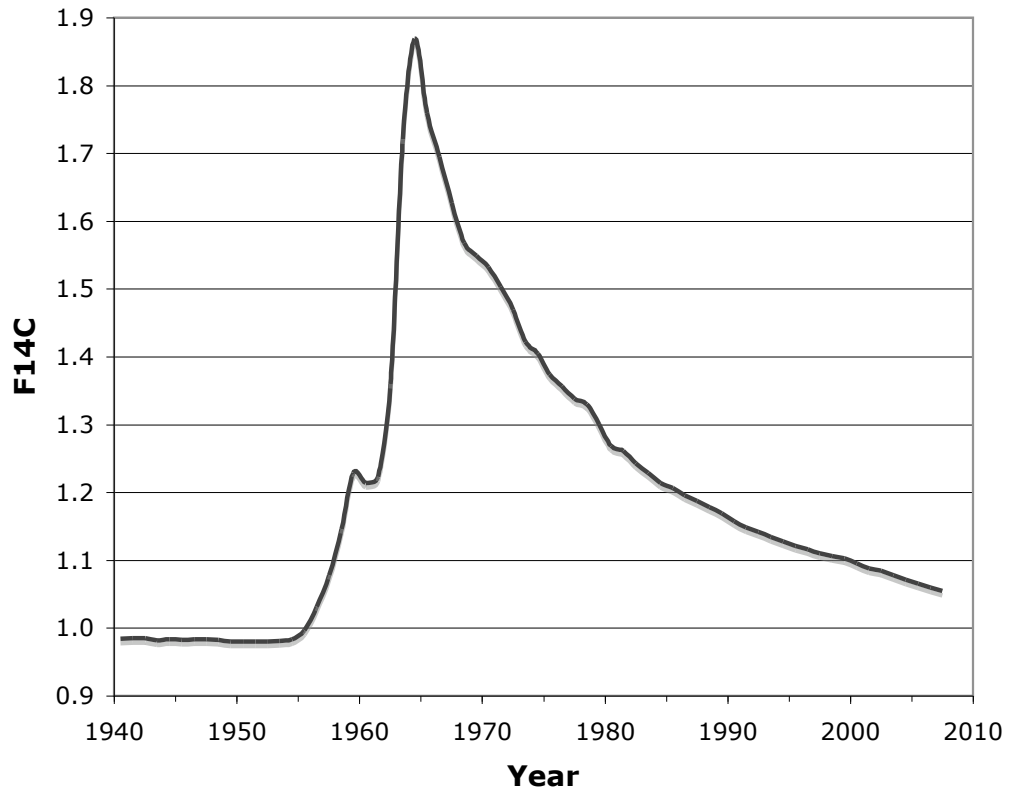


Figure 1. Northern hemisphere growing season average of atmospheric ^{14}C concentration in CO_2 from 1940-2007. The vertical axis uses the $F^{14}\text{C}$ nomenclature defined by Reimer, et al. (2006) specifically for bomb-pulse applications [11]. Other nomenclatures for expressing ^{14}C concentration can be found in the literature [12].